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Testing whether early diagenesis of skeletal carbonate is different in non-marine settings: contrasting styles of molluscan preservation in the Upper Jurassic of Portugal

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Abstract

This study tests the hypothesis that the early diagenesis of aragonite shells should differ fundamentally between marine and freshwater environments. This is predicted to be the case because aragonite is highly susceptible to dissolution in the TAZ (Taphonomically Active Zone) in low energy marine settings due to acidity caused largely by the oxidation of H₂S generated by sulphate-reducing bacteria, but reduced sulphide activity in freshwater settings should result in less early dissolution of aragonite. To test this hypothesis a range of fresh-brackish-hypersaline and marine limestones were sampled from the Upper Jurassic (mid Oxfordian) Cabaços Formation of central western Portugal. In these freshwater and brackish deposits, molluscs are preserved mostly as sparite shell replacements indicating that the original aragonite was preserved through the TAZ and was later replaced during subsequent burial by calcite cement. In limestones deposited in more marine to hypersaline settings, molluscan remains mostly consist of the calcitic layers of bimineralic bivalves, as shell where the original was wholly calcitic, or as gastropod steinkerns. Exceptions occur and reflect other factors such as higher energy conditions during deposition whereby organic matter, as the drive for microbial decay processes, was removed. The implications for molluscan preservation including some hydrocarbon reservoirs are discussed.

Key words: taphonomy; aragonite dissolution; non-marine; coquinas; sulphate reduction zone

1. introduction

The mechanisms and effects of the mobilization of carbonate during shallow burial in marine waters have been appreciated for some time (e.g. reviews by Sanders, 2003; Cherns et al., 2011): calcium carbonate, especially the more soluble aragonite, is dissolved in the uppermost sediment layer largely as a result of acidity caused by the oxidation of H_2S , itself produced by bacterial sulphate reduction. In lower energy settings organic matter can accumulate in finer grained sediments, whether siliciclastic, carbonate or of mixed composition, and sources the microbially mediated decay processes that then drive skeletal carbonate dissolution, and re-precipitation.

While most of the effectively syndepositionally dissolved carbonate back-fluxes to the water column, a fraction is re-precipitated as calcite in the sediment column in areas of increased alkalinity, such as depths where sulphate reduction takes place, to produce diagenetic bedding (Munnecke and Samtleben, 1996; Westphal and Munnecke, 2003; Wheeley et al. 2008). This oxidized zone, effectively the taphonomically active zone (TAZ) of Aller (1982) and Davies et al. (1989), will be controlled by diffusion from the overlying water column, if oxygenated, and by mixing caused by bio-irrigation (mainly burrowing).

The hypothesis being tested in this study is that as oxidation of H_2S is a major factor triggering dissolution of aragonitic shells, its effects should be less in many non-marine low-sulphate settings such that aragonitic shells may have a greater likelihood of entering the burial realm below the TAZ than in marine waters. This may seem a counter-intuitive argument as many marine waters, especially tropical and sub-tropical ones are less likely

than freshwaters to be undersaturated with respect to aragonite. However, the concentration of sulphate in freshwater ranges from ~10 to >500 μM , much lower than in seawater (28 mM) (Holmer and Storkholm, 2001).

To test this hypothesis we have examined a range of fresh–brackish–marine and hypersaline limestones from the Upper Jurassic Cabaços Formation of central western Portugal (Figs 1A, 2). While the data set shows a general difference in the preservation of former aragonite-bearing molluscan material between freshwater and other more saline waters, other factors need to be considered such as rates of burial, organic-matter content of the original sediment and the life position of aragonitic organisms relative to the TAZ.

1.1. Molluscan shell taphonomy

In low-energy marine settings, where organic matter can accumulate in the sediment and trigger a range of microbial processes and other reactions, aragonite under-saturation can lead to significant loss of shell material (e.g. Cherns and Wright, 2000; Cherns and Wright, 2009; Wright et al., 2003). Many mollusc shells are aragonitic, some are bimineralic (layers of calcite and aragonite) and some are entirely calcitic. The resulting fate of these shells during passage through the TAZ depends on their mineralogy and on their residence time in the zone of aragonite under-saturation (e.g. Sanders, 2003; Cherns et al., 2011). Aragonitic shells are preferentially dissolved and the resulting moulds are largely destroyed by bioturbation. Rarely steinkerns representing internal sediment fills of such shells can be preserved. Complete micritisation of the aragonite shell leads to its replacement by calcite but it is then difficult to distinguish such shell material from other forms of micritised, originally calcitic bioclasts. If the aragonite bypasses the TAZ into the final burial zone (FBZ; Olszewski, 2004), such as in the taphonomic windows identified by Cherns et al. (2008),

aragonite can persist or be congruently dissolved leaving a mould which can remain as a pore, or be filled by calcite cement, or be incongruently replaced (neomorphosed) to retain some palimpsest shell structure. Bimineralic shells retain their thin outer calcitic layer but the aragonitic layer(s) undergoes the same possible pathways as for wholly aragonitic shells. The remnant calcitic shell layer is commonly very thin and resembles the filaments described from various marine successions. Wholly calcitic mollusc shells generally survive little altered mineralogically into the FBZ.

2. Cabaços Formation

The Cabaços Formation (mid-Oxfordian; e.g. Ramalho, 1981; Azerêdo et al. 2002a,b; Kullberg and Rocha, 2014) crops out extensively in the Mesozoic Lusitanian Basin in west central Portugal (Figs 1, 2) (e.g. Wilson, 1979; Leinfelder and Wilson, 1998; Azerêdo et al., 2002a,b). It typically ranges from ca. 40-150 m in thickness at outcrop (but locally subsurface data point to 180-200 m or more) and disconformably overlies marine Middle Jurassic units. The three most representative of these are the mid-outer ramp Cabo Mondego formation (an informal unit) sensu Azerêdo et al. (2003), corresponding to the distal marine facies associations broadly named "Brenha" in industry reports, and the inner ramp Santo António-Candeeiros and Serra de Aire formations (Azerêdo, 2007), corresponding to the proximal marine facies associations broadly named "Candeeiros" in industry reports (Fig.2).

The basal Cabaços typically consists of medium to thick-bedded non-marine limestones, marls and lignites with a broad thickening trend towards the west and south from 12–16 m at Serra dos Candeeiros, Maciço Calcário Estremenho (Fig. 1B), where they are underlain by

c. 8-14m of pedogenic/black-pebble limestones and ferruginous marls (Azerêdo et al. 2002a, b), to over 20m at Pedrógão, to over 40 m at Serra d'El-Rei (Fig. 1B). In general it is typified by intercalations of marls, marly limestones, ferruginous limestones/marls and, locally, lignites and lignitic clays/marls, black-pebble limestones, and less commonly with pedogenic limestones (at the base), and isolated levels of microbial laminated micrites; deltaic sandstones and coral-oyster bioherms occur very locally, as at Cabo Mondego (Wright, 1985; Azerêdo et al. 2002b). Desiccation polygons up to 0.3 m in diameter are present locally and ferruginized irregular surfaces cap some beds such as at Pedrógão. In addition irregular fenestrae and vadose cements occur but are not widespread. Poorly preserved porostromate tubes are found in mm–cm grey calcimudstones to floatstones and at some localities, the lumps constitute small to large (several centimetres) oncoids. The palaeobiota is typified by non-marine taxa, in particular highly abundant charophytes and ostracods, common gastropods, bivalves, fossil wood remains, diverse palynomorphs of dominant continental source and rare reptile teeth. Extensive bioturbation has occurred at some levels.

The upper part of the Cabaços Formation typically consists of thin-bedded (locally medium-bedded), heterogeneous non-cyclic limestones, marly limestones, microbial laminites at places bituminous, and evaporites (Azerêdo et al., 2002b). Within the basin the total thicknesses in outcrop are extremely variable from as little as 15 m at Vale de Ventos, to at least 30 m at Pedrógão, 60m at Cabo Mondego but, in the Benfeito-1 well (Montejunto region), it reaches over 100 m (Fig. 1B). Thus, it is difficult to pick out a clear lateral thickening/thinning trend. In general the limestones consist of bioclastic calcimudstones to packstones, with minor bioclastic grainstones which can be indistinctly to clearly graded,

with parallel and low-angle cross-lamination and may show erosive basal surfaces developed on the underlying micrite-dominated levels. Rare oolitic-intraclastic grainstones also occur. Desiccation cracks are common and dinosaur trackways also occur. Ostracods, including as coquinas, are abundant as well as charophyte gyrogonites and less common stems. Gastropods and bivalves are very common, associated with dasyclads (at places abundant), rare agglutinate-walled foraminifers, serpulids, very rare echinoid remains and lower-diversity palynomorphs. Bioturbation effects are common. Gypsum crystal pseudomorphs and nodules are common, typically calcitised or silicified, and rare metre-scale evaporite breccias occur. Metre-thick beds of anhydrite are recorded in the subsurface (namely, at Benfeito-1 well). The organic-rich deposits of the Cabaços Fm., corresponding to different lithologies (lignites, lignitic clays/marls, bituminous marls/marly limestones and bituminous microbial laminites), have yielded heterogeneous values for geochemical parameters, namely TOC, across the basin. For instance, Silva et al. (2014) record TOC values ranging 0.26–4.67 wt% at Pedrógão, 0.33–30.56 wt% at Cabo Mondego (material heavily impregnated with hydrocarbons) and 0.43–11.64 wt% at Vale de Ventos, whereas Spigolon et al. (2011) indicate 2.87–4.93 wt% (outcrop samples) and Gonçalves et al. (2015) <1 wt% (borehole samples) at different places in the Montejunto region. The palynofacies is also variable but overall data show that the particulate organic matter is mostly of continental origin, punctuated by minor events of marine-influence, which increases towards the upper part of the unit (Barron and Azerêdo, 2003; Spigolon et al., 2011; Silva et al., 2014; Gonçalves et al., 2015).

This study focuses on the Pedrógão section (Fig. 1B), which is one of the most representative and well documented outcrops of the Cabaços Formation, for which a detailed palaeontological framework is already available.

2.1. Pedrógão Section

The well exposed coastal section at Pedrógão beach (Fig. 1B) exhibits an Upper Callovian-Oxfordian succession that has been described in detail by Azerêdo et al. (2002a, b) and further addressed in several thematic works on microfossil groups, namely ostracods (Cabral et al., 1998; Azerêdo et al., 2002a; Pais et al., 2016), charophytes (Grambast-Fessard and Ramalho, 1985; Pereira et al., 1998, 2003; Azerêdo et al., 2002a) and palynomorphs (Barron et al., 1999; Barron and Azerêdo, 2003). Azerêdo and Cabral (2004), using a range of palaeontological (chiefly the ostracod associations) and sedimentological data, identified in the Oxfordian deposits four facies/palaeoecological subdivisions, which are here labelled Units 1-4, updated and taken as independent background to test the facies interpretations of the present study (Fig. 3).

Note however, that ostracod assemblages were generally recovered from marls which allow the removal of whole carapaces, whereas most mollusc samples allowing determination of shell preservation were taken from indurated limestones, thus a direct comparison between these sample sets is not possible. The emphasis is therefore placed on trends in terms of salinity and preservation rather than identifying the exact conditions for specific horizons. At Pedrógão the more thickly bedded lower unit (Unit 1) is overlain by thin

bedded upper Cabaços which can be sub-divided into three sub-units (Units 2-4; Azerêdo and Cabral, 2004).

2.1.1. Unit 1

The lowermost Cabaços, 24 m thick was designated as Unit 1 by Azerêdo and Cabral (2004) (Fig. 3), and is characterised at outcrop by mainly matrix-dominated limestones, with lignites and lignitic marls. Evidence of exposure is not common and includes desiccation cracks at three levels, rare fenestrae and possible evidence of pedogenesis at one bedding surface.

The fauna contains a range of mainly freshwater to slightly mesohaline ostracods, documented in detail by Azerêdo and Cabral (2004) such as the genera: *Theriosynoecum*, *Darwinula/Alicenula*, *Mantelliana*, *Sinuocythere*, *Timiriasevia* and *Klieana*. The ostracod fauna generally increases in diversity towards the top of this unit, almost always with the genus *Theriosynoecum* dominant, but there are clear variations in the dominant species (of *Theriosynoecum*, *Darwinula*, rarely *Mantelliana* or *Sinuocythere*), as well as ornamentation styles and abundance. The most common are: *Theriosynoecum* spp., indicating freshwater to slightly mesohaline conditions (Carbonel et al., 1988), found in assemblages as the dominant species, typically very abundant, exhibiting ornamentation variations; *Darwinula/Alicenula* spp., occur also in assemblages with one species dominant and very abundant, as very large forms, indicating slightly more salinity-tolerance than *Theriosynoecum*, but still freshwater to weakly mesohaline conditions as *Darwinula* can

occur today at salinities of possibly over 10 ‰ (Carbonel et al., 1988). The living species *D. stevensoni* (Bradley and Robertson) tolerates an increase in salinity up to a maximum of 15‰ (Hiller, 1972, in Meisch, 2000). *Mantelliana* spp., becomes more common upwards in the unit, indicating fresh?/brackish, meso- to polyhaline conditions, possibly tolerating salinities up to 30‰ (Carbonel et al., 1988). Starting from 14.55m above the base of the section, there are a few levels of small-sized, smooth or almost smooth specimens of the euryhaline *Sinuocythere pedrogaensis* Cabral and Colin (Colin et al., 2000; Azerêdo and Cabral, 2004) (*cf.* palaeocology in Colin et al., 2000). In addition *Timiriasevia* and *Klieana*, both mainly oligohaline forms (Carbonel et al., 1988) are present at only a few levels.

An abundant and diverse charophyte flora is present (Grambast-Fessard and Ramalho, 1985; Pereira et al., 1998, 2003; Azerêdo et al., 2002a, b), including horizons of *in situ* calcite-encrusted stems indicating former presence of *Chara* meadows (Fig. 4A). There is a diverse palynomorph assemblage (namely *Corollina cf. torosus* and spores related to the Bennettitales and Cycadales), together with the typically freshwater alga *Botryococcus*, with minor marine-influenced contributions (Barron et al., 1999; Barron and Azerêdo, 2003). The palynofacies is mostly of a terrestrial origin with sporadic marine influence with phytoclasts and amorphous organic matter (AOM) increasing/decreasing oppositely as the result of periods of lower/higher environmental restrictions and preservation conditions (Silva et al., 2014). Remains of fully marine forms are very rare.

Former aragonitic molluscs, mainly gastropods (locally abundant small-sized forms), are preserved predominantly as sparite shell replacements lacking any remains of wall structure (Fig. 4B). Bimineralic shells show a well-preserved outer, thin calcitic layer with a sparitic

former aragonitic layer (Fig. 4C). Thin (0.1mm) calcitic laminae of bimineralic forms (here referred to as filaments) also occur at several levels but are not abundant and are typically highly comminuted.

In summary, this lowermost unit in the Cabaços of Pedróvão has been interpreted as a mainly freshwater deposit, representing more or less permanent shallow coastal lakes (Azerêdo et al., 2002b), with vegetated fringes and/or marshes. The ostracod fauna generally increases in diversity towards the top of this unit, but almost always with the genus *Theriosynoecum* dominant, but the more common presence of *Mantelliana* and of *Darwinula* suggesting a shift towards slightly more salinity-tolerant populations. The appearance of *Sinuocythere pedrogaensis* is further evidence for episodes of slightly higher salinity.

Preservation of former aragonitic mollusc shells as sparite replacements indicates that the original aragonite survived through the TAZ and was later replaced during subsequent burial and congruent dissolution by calcite cement. Bimineralic shells that show a well-preserved outer, originally calcitic layer and the sparitic former aragonitic layer(s) (Fig. 4C) are also evidence that aragonite survived the TAZ. These bivalves tolerate oligohaline but not freshwater conditions (Fürsich 1981; Fürsich et al. 2009). The presence of some filament shell laminae, here interpreted as calcitic layers of bimineralic bivalves left after dissolution of aragonite laminae implies that some early dissolution of aragonite is likely to have occurred locally. The filament material is highly comminuted and would have been readily transportable and could be allochthonous. It is noteworthy that the ostracods from marls immediately below or above limestones with higher proportion of calcitic filaments also

show small-scale variations in faunal composition pointing to slight salinity increase (within the 'larger-scale' freshwater/brackish assemblages). Although this may be circumstantial evidence, it supports the possible occurrence of very short flooding episodes, bringing more saline water and with it transported, reworked filament material.

2.1.2. Unit 2

The nearly 8 m thick Unit 2 (Fig. 5) has predominantly matrix-rich limestones with some grainstones. Microbial laminites occur, and desiccation cracks are present at eight levels, some associated with fenestrae. Nodular textures occur at some levels forming conglomerate-like bed surfaces and irregular micro-cracking which resemble pedogenic alteration as described from the Cabaços Formation at Vale de Ventos, Serra dos Candeeiros (Azerêdo et al., 2002b), (Fig. 1B). Ostracod faunas, in contrast to previous unit, are dominated by the euryhaline species *Sinuocythere pedrogaensis*, as small specimens, smooth but also slightly reticulated, suggesting some ecological change, although the mainly oligohaline *Theriosynoecum* and some other genera present in Unit 1 also occur. A new species belonging to a genus close to *Klieana* (*Klieana*? sp.) appears, probably more euryhaline, due to its morphological characteristics, as suggested by the almost complete absence of median vertical sulcus; overall, the assemblage suggests brackish to slightly more saline affinities.

Charophyte gyrogonites while ubiquitous are less abundant, stems are rare, and thin horizons packed with stems of the dasyclad *Barattoloporellopsis lusitanica* (Ramalho), formerly *Heteroporella lusitanica* (Ramalho) (see Granier et al., 2017) occur. Benthic

foraminifera such as *Kurnubia palastiniensis* Henson and other agglutinate-walled forms occur at some levels (Azerêdo et al., 2002a; Azerêdo and Cabral 2004). The palynofacies shows assemblages of continental and less common marine contributions, namely extremely rare freshwater palynomorphs, *Botryococcus* and dinoflagellate cysts, and high amounts of AOM (Barron et al., 1999; Barron and Azerêdo, 2003; Silva et al., 2014).

Thick shelled *Isognomon rugosus* oysters (Fursich et al. 2009) form shell concentrations associated with serpulid buildups (Fig. 5A, B). The most common fossils are calcitic shells of *Praeexogyra pustulosa* oysters in shell pavements, and there are horizons with filaments of bimineralic bivalves. Another shell horizon above a desiccation cracked surface has concentrations of neomorphosed shells of shallow burrowing nuculoid bivalves and small gastropods. Fursich et al. (2009) and Fursich (1981) interpreted autochthonous *Isognomon* banks as indicating a restricted salinity environment, while *P. pustulosa* could tolerate reduced salinity brachy- to mesohaline conditions. Formerly aragonitic gastropod shells are sparite-replaced or, less commonly, neomorphosed; pyrite in a sparite replaced and infilled shell may indicate pyrite growth during or after sparite growth (Fig. 5C).

In summary, the evidence of frequent exposure and microbial laminites, with a few grainstones, suggests fluctuating energy levels of a very shallow water-body. There is presence of a limited range of more marine-influenced indicators such as forams (agglutinated forms, which range from average-marine to lower salinities) and dasyclads (typical of marine lagoonal settings, from near normal-marine to more restricted conditions; in particular, *B. lusitanica* is found also at very restricted, brackish lagoon facies of the Cabaços Formation in the Vale de Ventos, Serra dos Candeeiros region (Fig. 1B), which

suggests it was more tolerant to slight salinity fluctuations than usually acknowledged for dasyclads (Azerêdo et al., 2002a). The bivalves indicate restricted, brachy- to mesohaline salinities. The aforementioned markers are coupled with less common non-marine indicators such as charophyte stems/gyrogonites and changes in the ostracod biota, suggesting fluctuating salinities of a brackish setting, with at times a more marine influence.

Molluscan remains (Fig. 5) consist mainly of calcitic shells or shell layers, and sparite replacements or less common neomorphic replacements of aragonite by calcite. The latter indicate that the original aragonite was preserved through the TAZ, being later replaced during subsequent burial and congruent dissolution by calcite cement. The preservation of the former aragonitic dasyclad *Barattoloporellopsis* also supports this interpretation but the sparite crystals do not exhibit a pattern of pore-infilling precipitated material, therefore reinforcing the assumption that they are a neomorphic replacement of the original aragonitic coating of the dasyclad (Granier et al. 2017). Minor occurrences of other neomorphically replaced former aragonitic shells also suggest that some aragonite passed through the TAZ. The presence of some pyrite within the sparite-replaced walls and sparite infill of the body chambers in some gastropods (Fig. 5C) suggests sulphate reduction took place but probably after the aragonite had passed through the TAZ.

2.1.3. Unit 3

This unit, some 6 m thick (Fig. 6), consists predominantly of matrix-rich limestones, commonly fossiliferous, interbedded with a few laminites, thin marly layers and evaporites at three levels; evidence of exposure is weaker, though rare fenestrae and two pedogenic

horizons occur. Porostromate textures, including reworked examples are present, together with coated grains, reworked microbial structures, microbial boundstones and peloidal-intraclastic lenses.

The ostracod assemblage is of lower diversity and indicates variably brackish to saline conditions, as it lacks the oligohaline *Theriosynoecum*, whereas the euryhaline *S. pedrogaensis* is the dominant form and showing differences in size and carapace surface features. The fresh/brackish form *Darwinula* occurs, and locally the euryhaline marine genus *Macrodentina* (*Galliaecytheridea*? in Azerêdo and Cabral, 2004), together with *Klieana*? Dasyclad remains are less common but a low diversity charophyte gyrogonite assemblage is present (Pereira et al., 2003; Azerêdo and Cabral, 2004), whereas charophyte stems are rare. *Botryococcus* while present is absent towards the top of the unit (Barron et al., 1999; Barron and Azerêdo, 2003). The foram *Pseudocyclamina* and a few other agglutinated forms occur and rare echinoid remains are found.

A particularly prominent, c. 30 cm thick shell-bed exhibits abundant spar-replaced shells of deep-burrowing aragonitic bivalves (*Pholadomya*) in imbricated concentrations and in situ (Fig 6A) with bioclasts forming either planar or cross laminations, the latter mainly towards the top of the bed. Shelly material makes sub-layers within the bed, with sharp erosional or gradational bases. Many of the shells were apparently exhumed and reworked, redeposited and rapidly buried locally, with little fragmentation, but associated in situ shells suggest this was followed by some recolonization.

Unit 3 shows a marked range of salinity indicators such as forams, rare echinoid debris, coupled with the dasyclads, thus suggesting restricted lagoonal marine conditions. The shell beds are interpreted as the product of storm events.

Overall, aragonitic molluscs preserved by sparite replacement are uncommon (Fig. 6A, B) although exceptions include one bed rich in very small gastropods close above an evaporite horizon, and the imbricated shell horizon (Fig. 6A). Filament material is less rare, although not common (Fig. 6C).

2.1.4. Unit 4

Unit 4 (20 m thick) includes prominent microbial laminites (Fig. 7A), often bituminous and evaporite layers. Oolitic/intraclastic packstones and grainstones are present at the top of the section. There is evidence of some pedogenesis, represented by nodulization fabrics, circumgranular and irregular dissolution/brecciation cracks, and rare very thin carbonate crusts resembling calcicutans.

This unit has most abundant ostracods, like the previous units, but almost exclusively of the euryhaline form *Sinuocythere pedrogaensis* with larger sized and reticulate carapaces (Colin et al., 2000; Azerêdo and Cabral, 2004). This species even forms coquinas within laminites and is locally closely associated with evaporites. Forams are more common including *Pseudocyclamina parvula* Hottinger, which is known to increase upwards into the overlying marine Montejunto Formation (Ramalho, 1981; Azerêdo et al., 2002a, b). Rare echinoid fragments occur also. Dasyclads are rare and disappear near the base of the unit;

charophyte remains are less abundant, more so upwards and show the lowest diversity (Pereira et al., 2003), and the palynomorphs are of lower diversity and marine influenced (Barron and Azerêdo, 2003). Typical marine trace fossils occur locally at the lower part of the unit and include *Rhizocorallium* and *Thalassinoides*. Rare echinoid fragments occur also. Molluscan shell material is predominantly of filaments with occasional steinkerns of small gastropods (Fig.7B) and rare neomorphosed aragonite shells.

This unit is interpreted as representing shallow, margino-littoral deposition under highly variably hypersaline to marine-influenced conditions, becoming less hypersaline upwards. The lack of sparitic preservation suggests former molluscan aragonite did not survive the TAZ but the filament debris indicates their former presence. This may also explain the decline on preservation of the dasyclads although that could also reflect ecological factors.

2.2. Taphonomic interpretation

The Pedrógão succession shows a broad relationship (Fig. 8) whereby former aragonitic shells were initially preserved in fresh to brackish settings from early dissolution in the TAZ but dissolved and filled with calcite cement during later diagenesis (Units 1 and 2). This applies to bimineralic forms as well as those with a wholly aragonitic original shell. Sparite replacement preservation declines through Unit 3, where evaporites become more common. In Unit 4, which represents marine to hypersaline settings the aragonite was preferentially removed syndepositionally leaving only originally calcitic forms, the filaments of calcitic layers of bimineralic forms or steinkerns. However, the pattern is not always consistent. Thin filaments representing only the calcitic layers of bimineralic forms are also

common in some beds in Unit 1, suggesting that some aragonite dissolution had occurred, but the filament material is generally seen as very small fragments which could have been transported in from other settings. Overall there appears to have been a reduced level of early, syndimentary dissolution compared with the overlying units. The former aragonitic dasyclad *Barattoloporellopsis* is also most commonly found in Unit 2, suggesting it was tolerant of lower salinities like some extant dasyclads but its rarity in Units 3 and 4 might largely be an ecological not taphonomic effect.

Additional factors influence the survival of aragonitic shells past the effects of the TAZ leading either to their continued preservation as aragonite, or to the replacement of aragonite by calcite, as reviewed by Cherns et al. (2008). For example, in high energy deposits including coquinas, fine organic matter is less likely to accumulate, when coupled with the likelihood of more oxygenated sediment, this reduces the amount of sulphate reduction and hence of aragonite dissolution due to sulphide oxidation. A common situation where aragonite can be preserved from early dissolution is in event (storm) beds where rapid burial emplaces the aragonite below the TAZ and many such examples are known (Cherns et al., 2008). This may explain the preservation of the former aragonitic bivalves in Unit 3 which appear imbricated and may have been reworked and rapidly buried (Fig. 6A). Some of these shells are in life position and deeper burrowing forms may be predisposed to have the aragonite initially preserved as they live below the main part of the TAZ (Wright et al., 2003). De Francesco et al. (2013), from assemblages in Holocene lakes in Argentina, ascribed better preservation of molluscs from brackish rather than freshwater lacustrine settings to differences in calcium carbonate saturation. Whether shell thickness played a role in the differential preservation across the salinity gradient is not possible to

determine as the aragonitic component in the marine-hypersaline units has been removed. Another consideration is that the availability of Fe in the more argillaceous facies (such as much of Unit 1) might have led to a reduction in dissolution in the TAZ (e.g. Kidwell et al., 2005).

The availability of Fe is evidenced by the fact that pyrite is found throughout the four units although is highly variable in abundance. However, sparitic mollusc shells are commonly present in the less argillaceous limestones in Units 1 and 2.

With the evidence of a significant difference in early aragonite preservation having affected the Cabaços Formation at Pedróvão, and reflecting salinity changes and marine influence, a likely explanation is that sulphate availability was a critical factor. The concentration of sulphate in freshwater can be three orders of magnitude lower than in seawater (Holmer and Storkholm, 2001), limiting the potential production of H₂S as the main agent for triggering aragonite undersaturation and dissolution. This effect is somewhat more complicated as sulphate reducing bacteria, the presumed main source of H₂S in shallow buried sediments occur at similar concentrations in freshwater settings to marine ones (Jørgensen and Bak, 1991; Holmer and Storkholm, 2001). Sulphate usually only occurs to <10 cm into freshwater sediments (e.g. Cook and Schindler, 1983; Holmer and Storkholm, 2001), and as a result the zone with the most active sulphate reduction is found in the top 0.1 m of sediment (Ingvorsen et al., 1981; Sass et al., 1997), which is in contrast to marine sediments where sulphate reduction can be deeper, although sulphate reduction is often intense despite the low sulphate concentration (Holmer and Storkholm, 2001). Cristini and De Francesco (2017), from a study of Holocene lakes from Argentina, note that the top 10 cm of the sediment column is less favourable for molluscan preservation. Although data are

limited, they suggest this reduced thickness of the TAZ compared with marine sediment might relate to the role of deeper burrowers in marine settings. The depth of burrowing in lake sediments where bottom waters are not anoxic is much shallower than in marine systems (Hasiotis et al., 2012), generally within the uppermost 0.3 m compared with a maximum range of 1-4 m in marine settings. Thus compared with low energy marine settings the TAZ in lakes should be thinner with skeletal aragonite potentially having a shorter residence time in the TAZ than in a marine setting, making it more likely to survive the zone of aragonite dissolution, and also more readily buried below the TAZ during a storm event. This raises the question as to whether aragonite preservation is more sensitive to deposition rate in non-marine, low sulphate settings.

3. Discussion

This study highlights the possible significant preservational differences in molluscs between limestones from freshwater to marine settings, but also shows the range of other factors besides sulphate concentrations that could be involved. There are testable implications arising from this proposal that early diagenesis differs in low sulphate waters. Firstly, former aragonitic molluscs should be more commonly preserved in low energy, low sulphate settings, especially aragonitic shallow-burrowing infaunal bivalves, along with gastropods, which are typically those most readily removed from marine low energy deposits as a result of aragonite dissolution in the TAZ (Cherns and Wright, 2009). Related to this should be the preservation of juvenile forms and micromolluscs (Cherns and Wright, 2011). As carbonate released by skeletal aragonite dissolution is the source of the carbonate

forming diagenetic nodules and bedding, these might be less common in low sulphate settings if early molluscan aragonite dissolution is much reduced. However, this would also be the case with the alternative proposal (Munnecke and Westphal, 2005) that aragonitic mud is the main source of the diagenetic calcite since this would be less likely in freshwater settings where aragonite producers such as calcareous algae are absent and where precipitation directly from the water column is less likely in low salinity lake waters. During the early Palaeozoic the TAZ was also very thin and as a result the location of cementation below the TAZ was also very near the sediment surface resulting in the widespread occurrence of reworked cemented material as flat pebble conglomerates (Wright and Cherns, 2016). Similar conglomerates do not seem to be a feature of sub-littoral lake deposits and were not noted in the units described in this study.

As a consequence of the TAZ being thin, perhaps even <10 cm, in low energy lacustrine settings, rapid burial might favour more common aragonite preservation in thin event beds in such low sulphate settings. While it would be difficult to assess the relative frequency of event beds between marine and non-marine successions there are certainly examples of shell beds in non-marine successions. Radley (2009) provides many examples of thin shell beds preserved in Mesozoic brackish-to lacustrine successions, but favours “a fundamental biological control on their genesis; namely small shell size, high reproductive rates and adoption of conservative life modes” (p. 715), and the role of reduced sulphate levels in such settings is not considered. Pan et al. (2012) provide a detailed analysis of lacustrine shell beds from the early Cretaceous Yixian Formation at Sihetun, Beipiao, PRC, focussing on biological and stratinomic effects not on preservation.

Complex patterns of aragonitic molluscan preservation, including the survival of aragonite, are recorded from the late Jurassic-early Cretaceous 'Purbeckian' facies of southern England by El-Shahat and West (1983), representing brackish lagoons. Skawina (2013) provides one of the most detailed analyses of bivalve taphonomy in a lacustrine, charophyte-bearing succession, from the late Triassic of Poland, noting the disarticulated unionoids (originally aragonitic) valves showed no signs of dissolution, interpreting this as evidence the dissolution of the aragonite was a late diagenetic event.

The most extensive lacustrine molluscan coquinas, of early Cretaceous age, occur in the South Atlantic. These are documented from the offshore Brazil in the Coqueiros Formation (Lagoa Feia Formation) of Campos Basin (Carvalho et al., 2000; Thompson et al., 2015), the Itapema Formation of Santos Basin (Carlotto et al., 2017), and the Sergipe-Alagoas Basin in the Morro de Chaves Formation (Kinoshita, 2010; Tavares et al., 2015), and offshore West Africa in the 'Toca' (e.g. the Congo and Cabinda basins; Harris, 2000). In the Campos Basin the Barremian coquinas are associated with fluvial-alluvial deposits and lacustrine carbonate bodies, deposited in a complex lake system controlled by local environmental conditions and tectonics. Its thickness varies from 2,400 m in palaeostructural lows to 0 m in palaeostructural highs. The main hydrocarbon reservoir type consists of coquina lenses characterized by units over 10 m thick made up of amalgamation of relatively thin layers (0.20–1 m thick) of bivalve-rich calcirudites, classified as grainstones, that locally show cross stratification, associated with thin layers of matrix-poor and matrix-dominated carbonates and no siliciclastic rock intercalations. Successions comprised of this facies sub-association form layers 20–550 m thick. The sedimentary features suggest high energy and shallow water depositional conditions. Although many of the coquina deposits represent high energy shoreline facies others are associated with low energy matrices and the contrast

501 with their marine equivalents where infaunal, former aragonitic bivalves are only rarely
502 preserved might be explained by the lower sulphate content of the lake waters. However,
503 non-marine systems are highly varied and dynamic in terms of their chemistries and many other
504 diagenetic pathways are possible for molluscan skeletal preservation (Fursich and Pan, 2016).

505

506 **4. Conclusions**

507

508 A fundamental difference should exist between the early diagenesis of aragonite in
509 marine and freshwater environments because of the degrees of sulphide activity and its
510 effect on aragonite preservation. Growing evidence shows that aragonite is lost, effectively
511 syndepositionally, from low energy marine sediments because of undersaturation triggered
512 by oxidation of H₂S generated by sulphate-reducing bacteria. Such effects should be reduced
513 in sulphate-poor settings such as freshwater lakes. To test this hypothesis a range of fresh-
514 brackish-hypersaline and marine limestones was sampled from the Upper Jurassic (mid-late
515 Oxfordian) Cabaços Formation of central western Portugal. In limestones deposited in
516 freshwater to brackish settings many molluscs, mainly gastropods, are preserved as sparite
517 shell replacements indicating that the original aragonite was preserved through the TAZ but
518 were later replaced during subsequent burial by calcite cement. In limestones representing
519 deposition in more marine to higher salinity settings molluscan remains mainly consist of the
520 calcitic layers of bimineralic bivalves, or of calcitic shells, or gastropod steinkerns. While
521 there seems to be dominant diagenetic/taphonomic preservation patterns related to the
522 two main facies associations (fresh/fresh-brackish water and marine-hypersaline), a
523 consistent pattern is not always present, reflecting the high-frequency salinity changes
524 recorded in the Cabaços Formation. This potential preservational bias in low sulphate waters

525 towards aragonitic fossils requires further study and might explain the anomalous thickness
526 and abundance of some lacustrine molluscan limestones such as those making significant
527 hydrocarbon reservoirs in the South Atlantic.

528

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Figure captions

Fig. 1. A, inset map of the Iberian Peninsula indicating study area in western Portugal; B, simplified geological map of the onshore part of the Lusitanian Basin, western central Portugal showing outcrop area of Jurassic limestones, with location of Pedrogão and other key localities in bold.

Fig. 2. Simplified stratigraphical framework (not to scale) for the Middle–early Upper Jurassic of the Lusitanian Basin (Cabaços Formation shaded) showing the interpretation of dominant depositional environments/facies types.

Fig. 3. Log of the section at Pedrógão showing the stratigraphic distribution of sedimentary features, key biota and types of molluscan replacement. Note that while ostracods are ubiquitous in almost all levels, the associations are salinity-sensitive so composition and proportions of taxa differ (see text), which is summarized by marking distribution as two broad groups: i) freshwater/brackish and ii) euryhaline forms. When both groups occur, the dominant one is indicated by a square around the bullet. Four facies/palaeoecological subdivisions recognised by Azerêdo and Cabral (2004) from a range of palaeontological (chiefly the ostracod associations) and sedimentological data are here labelled Units 1-4, updated and taken as independent background to test our interpretations.

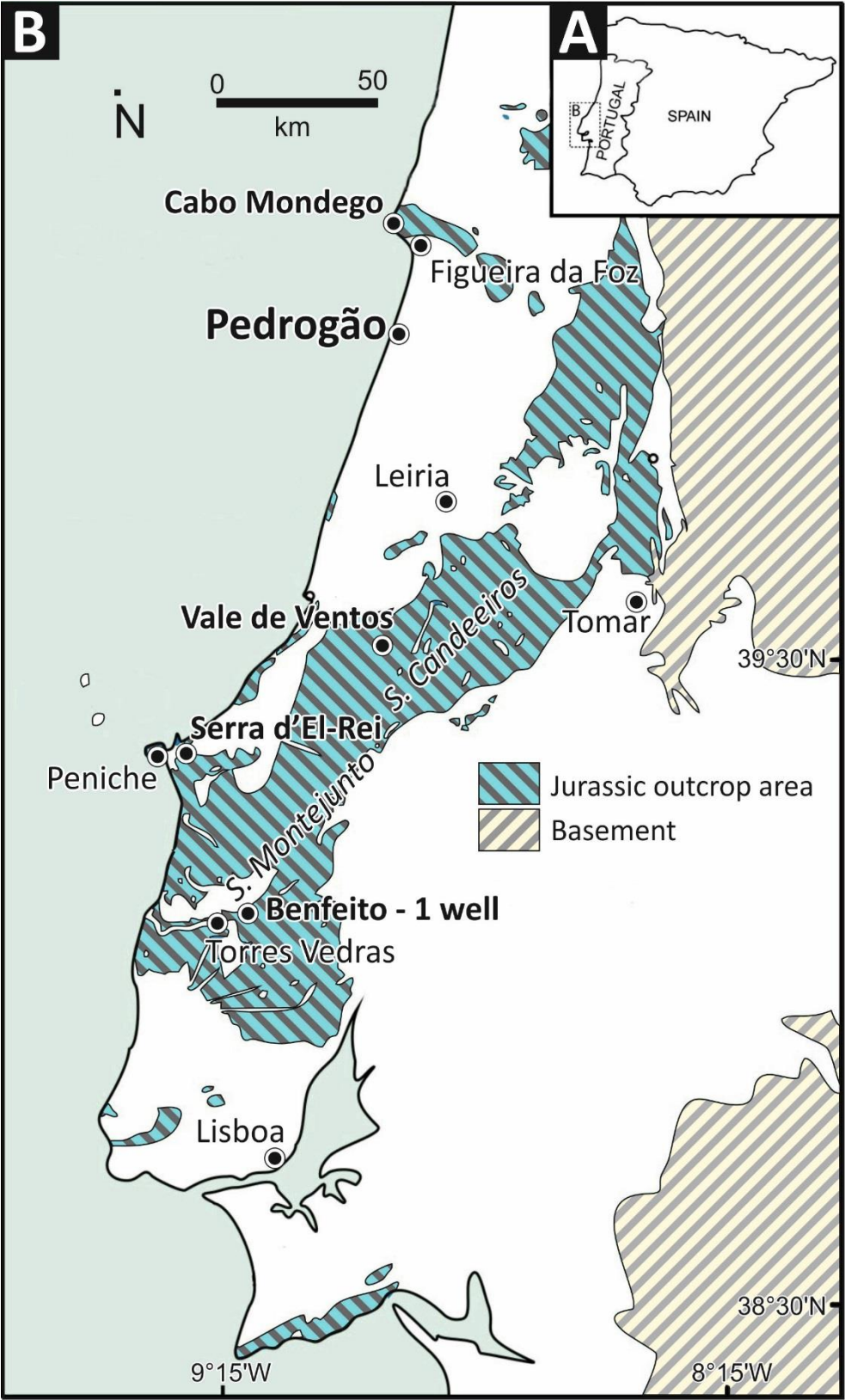
Fig. 4. Lithofacies Unit 1 at Pedrógão. A, calcite encrusted charophyte stems in coquina near the base of the Cabaços Formation; B, gastropod shells, some partially replaced by sparite and some with thin, well preserved calcitic outer layer (residue retrieved from >0.5mm fraction of a marl sample studied for microfossils); C, fragments of well-preserved outer calcitic layer of a bimineralic gastropod shell, and a well preserved calcitic ostracod carapace.

Fig. 5. Lithofacies Unit 2 at Pedrógão. A-B, *Isognomon* shell bed, shells mainly articulated, with well preserved calcitic outer shell layer; C, gastropod with sparite walls and infill of the body chamber including some pyrite, which suggests pyrite formation during or after sparite growth.

Fig. 6. Lithofacies Unit 3 at Pedrógão. A, imbricated pholadomyacean shells with sparite-replaced walls; B, sparite-replaced former aragonitic gastropod shell and *B. lusitanica* (dasyclad); C, 'filaments' representing calcitic outer layer(s) of bivalve shell, inner aragonitic layers have been dissolved.

Fig. 7. Lithofacies Unit 4 at Pedrógão. A, microbial laminites; B, steinkern of gastropod, suggesting early dissolution of formerly aragonitic walls, associated with ostracods and a charophyte gyrogonite.

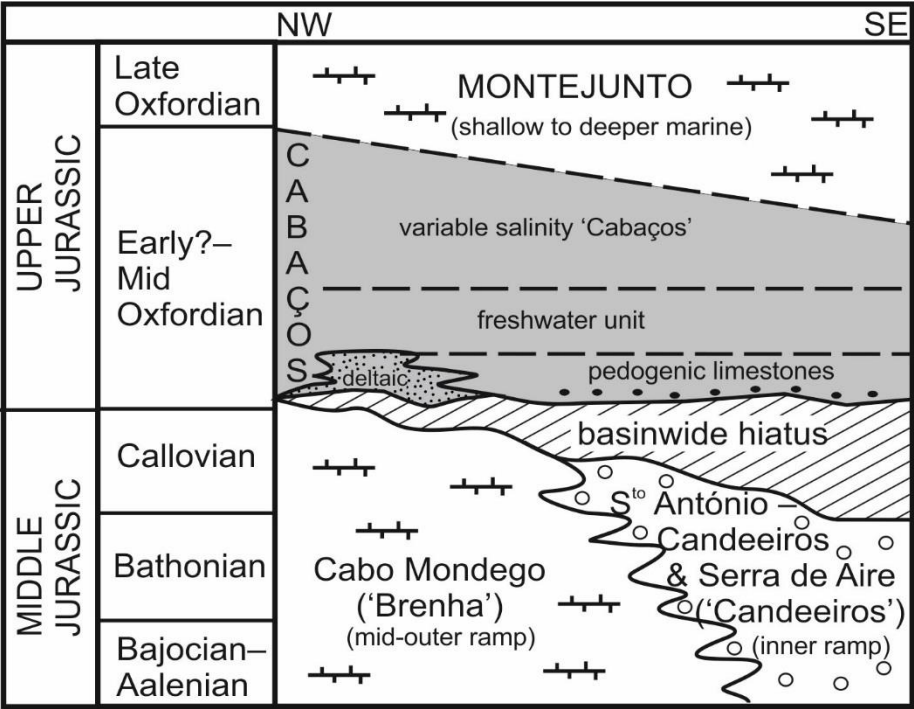
Fig. 8. The effect of salinity on the potential for aragonite shell preservation. In fresh to brackish (low sulphate) waters sulphide activity is limited in the TAZ (Taphonomically Active Zone), increasing the potential for initial aragonite preservation, later to be replaced by sparite in the FBZ (final burial zone). In more sulphate-rich waters aragonite is normally preferentially removed and moulds are typically destroyed by bioturbation; filaments representing the outer prismatic calcite shell layer of bimineralic bivalves and steinkerns may be preserved.



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754 Figure 2



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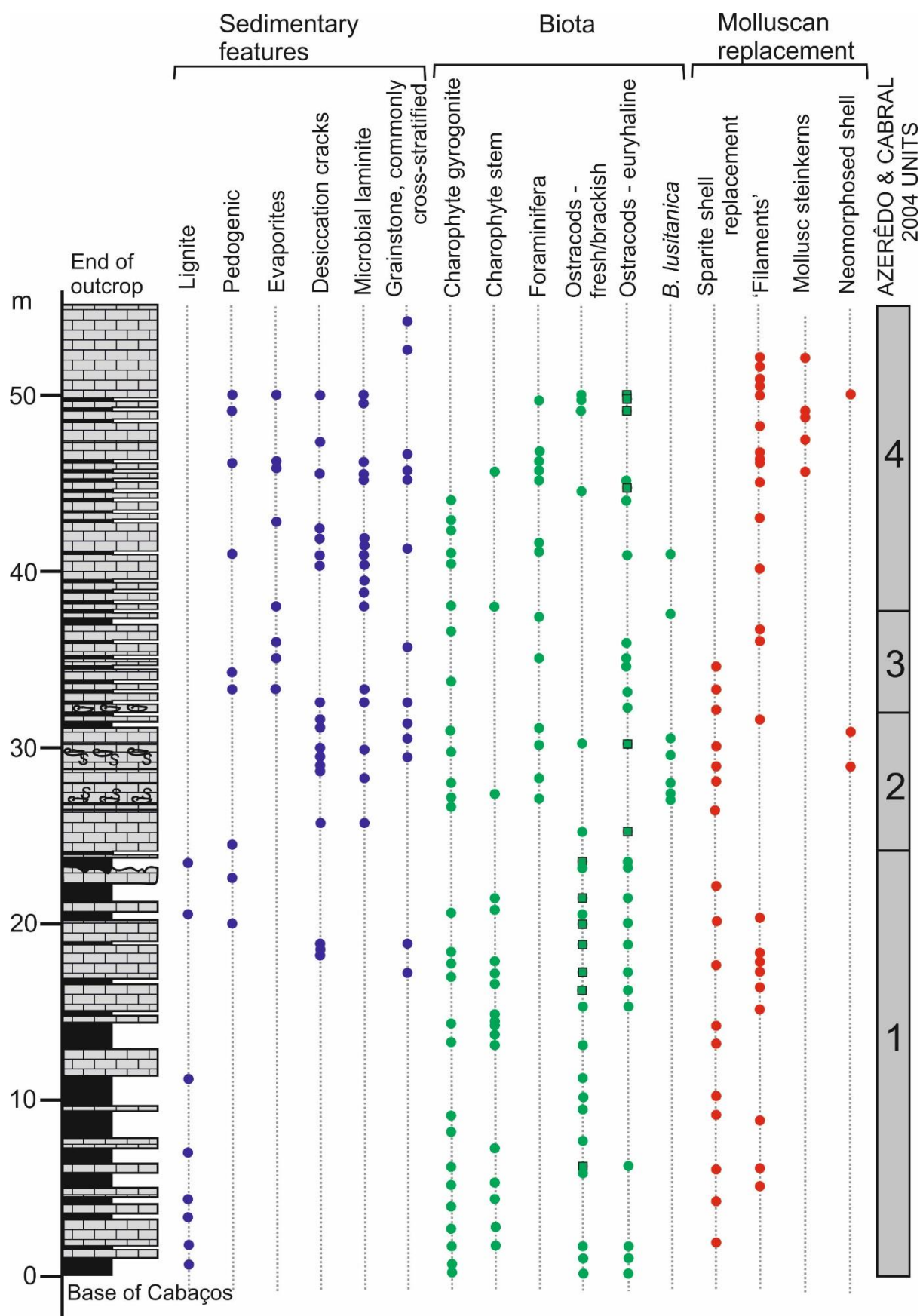
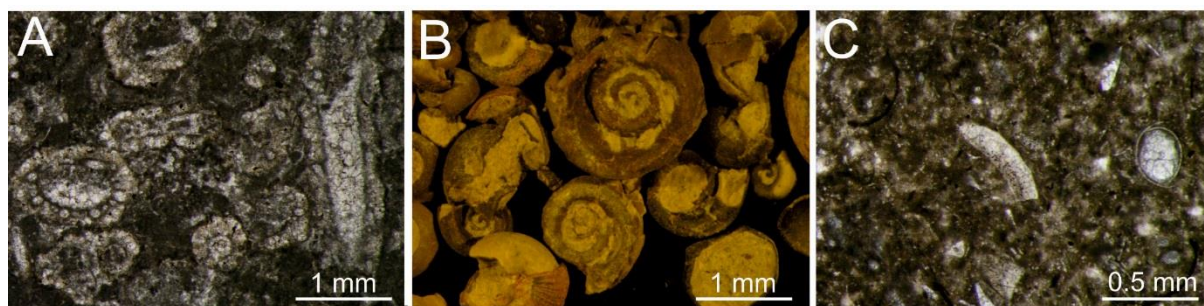
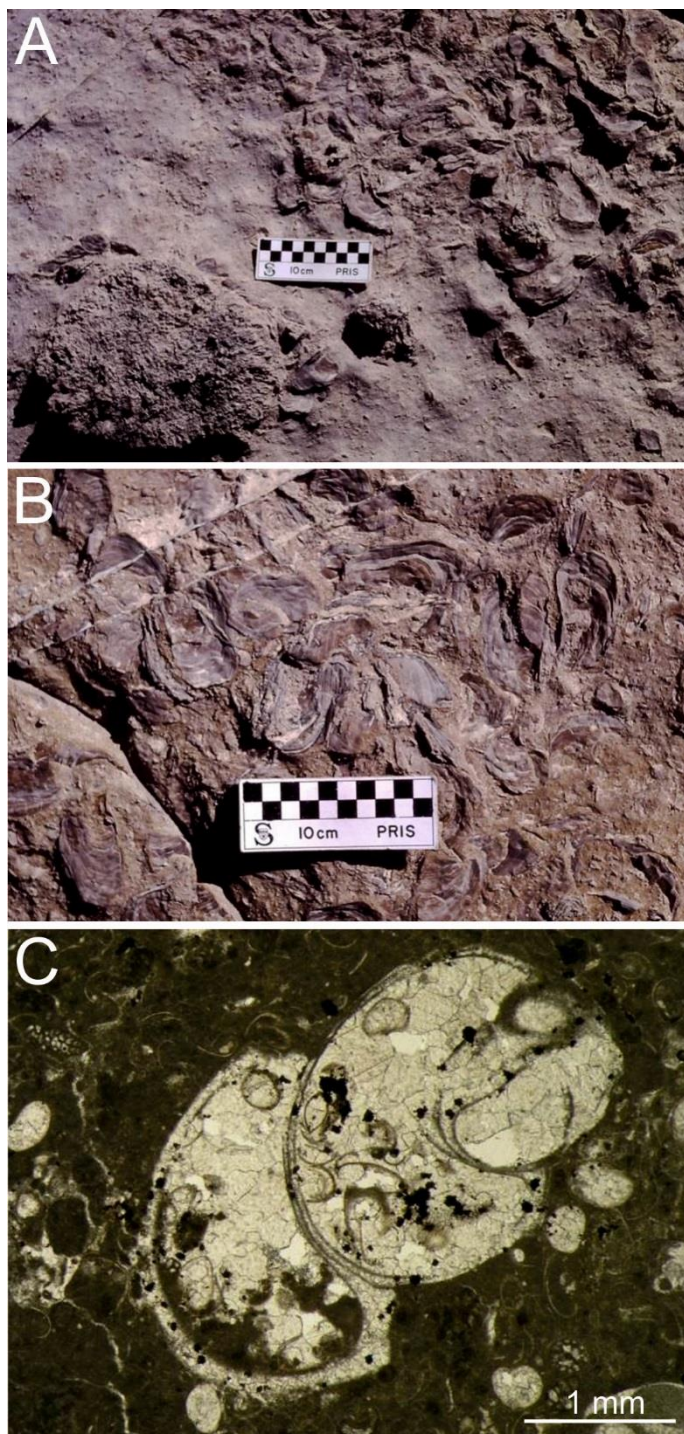


Figure 4



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778 Figure 5



779

Figure 6

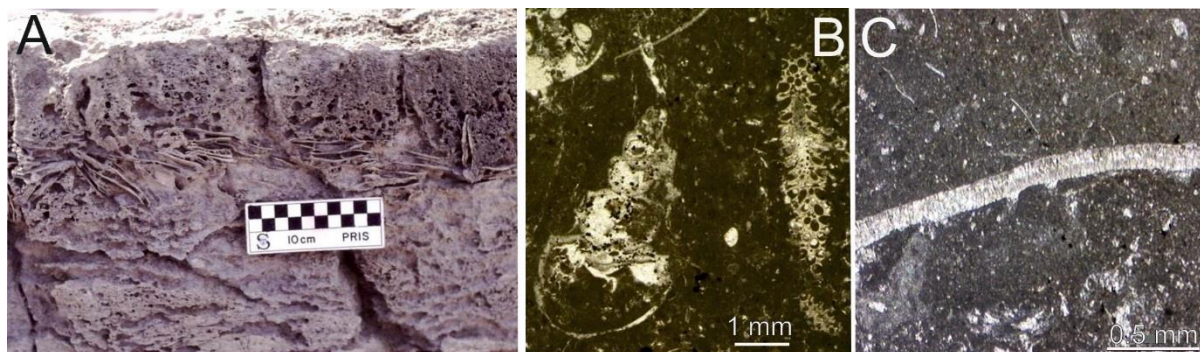


Figure 7

